Preprint
UCRL-JC-145338

Reducing Collisional Breakup of a System of Charged Particles to Practical Computation: Electron-Impact Ionization of Hydrogen

C.W. McCurdy, M. Baertschy, W.A. Isaacs, T.N. Rescigno

This article was submitted to
22nd International Conference on Photonic, Electronic and Atomic Collisions, Santa Fe, New Mexico, July 18-24, 2001

U.S. Department of Energy
Lawrence Livermore National Laboratory

August 24, 2001

REDUCING COLLISIONAL BREAKUP OF A SYSTEM OF CHARGED PARTICLES TO PRACTICAL COMPUTATION: ELECTRON-IMPACT IONIZATION OF HYDROGEN

C. W. MCCURDY[1,2]
M. BAERTSCHY[3], W. A. ISAACS[4], *AND* T. N. RESCIGNO[1,4]
[1]*Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., Berkeley, CA 94720, USA*
[2]*Department of Applied Science, University of California, Davis, Livermore, CA 94551, USA*
[3]*University of Colorado, JILA, UCB 440, Boulder, Colorado 80309, USA*
[4]*Lawrence Livermore National Laboratory, PO Box 808, Livermore, CA 94551, USA*
E-mail: cwmccurdy@lbl.gov

It has been a goal of researchers in the area of atomic collisions for nearly half a century to reduce to practical computation the simplest problem in collisional ionization: the electron-impact ionization of atomic hydrogen. The principal barrier to solving this problem has been the difficult boundary conditions that apply to the complete breakup of a system charged particles. We describe how this goal has been accomplished in the last five years by the application of the mathematical transformation of "exterior complex scaling" together with an appropriate formalism for computing the breakup amplitudes from a numerical representation of the complete solution of the Schrödinger equation. Some successes of other recent approaches to this problem are also described.

1 Introduction

By the end of the twentieth century essentially every feature of two-electron problems in quantum mechanics had for decades been reduced to practical computation. A problem has been reduced to "practical computation" when a formalism and the associated numerical algorithms have been developed that allow the calculation, with currently available computing capability, of the relevant physical quanties to any accuracy that can be tested by experiment. For example, the bound states of two-electron atoms in nonrelativistic quantum mechanics have been such a "solved problem" since the work of Hylleraas [1] and Pekeris [2]. The same is true of atomic collision processes that have one electron in the continuum (ordinary elastic and inelastic scattering) as a result of an extended effort in the 1970s and 1980s involving many investigators.

However, for collision processes with two electrons in the continuum this goal was only reached in the last years of the century, and that effort is what this lecture is about. The central difficulty that impeded progress on the problem of three-body breakup in Coulomb systems, particularly for the collisional breakup or "e,2e" problem (as opposed to double photoionization), is the cumbersome asymptotic form of the scattering wave function that the formal theory of ionization imposes. Peterkop [3] and Rudge and Seaton [4] deduced the appropriate boundary conditions in the 1960s to be

$$\Psi^{+}_{\rm ion}(\mathbf{r}_1,\mathbf{r}_2) \underset{\rho\to\infty}{\longrightarrow} -f_i(\hat{r}_1,\hat{r}_2,\alpha)\sqrt{\tfrac{i\kappa^3}{\rho^5}}\exp\{i[\kappa\rho+\frac{\zeta(\hat{r}_1,\hat{r}_2,\alpha)}{\kappa}\ln(2\kappa\rho)]\}, \qquad (1)$$

where f_i is the ionization amplitude and the hyperspherical coordinates are defined by $\rho=(r_1^2+r_2^2)^{1/2}$ with $\alpha=\tan^{-1}(r_1/r_2)$, and κ is related to the total energy by $E=\kappa^2/2$. The most obvious difficulty in applying this boundary condition is that the coefficient

$\zeta(\hat{r}_1, \hat{r}_2, \alpha)$ of the logarithmic phase depends on the distances and ejection angles of both electrons. However, the worst problem in applying Eq. (1) may be the fact that it is not separable in spherical coordinates, and therefore much more cumbersome to apply to numerical calculations which are per force done in that coordinate system. As a consequence, no one has yet applied Eq.(1) to the numerical solution of the Schrödinger equation for the ionization problem.

The formal theory of ionization poses another challenge to computation as well, and that is that the ordinary expression for evaluating the amplitude, starting from the scattering wave function that solves the Schrödinger equation, does not apply, because defined in the usual way it would have an infinite phase associated with integrating an expression with logarithmic phases over an infinite volume. Instead the amplitude is give by [3,4,5]

$$f(\mathbf{k}_1, \mathbf{k}_2) = -(2\pi)^{5/2} e^{i\Delta(\mathbf{k}_1, \mathbf{k}_2)} \iint \Psi^+(H - E)\phi(-\mathbf{k}_1, z_1)\phi(-\mathbf{k}_2, z_2) d\mathbf{r}_1 d\mathbf{r}_2 \qquad (2)$$

with *effective charges* in the one-body Coulomb functions, $\phi(-\mathbf{k}, z)$ depending on both the energy and direction of ejection of each electron,

$$\frac{z_1}{k_1} + \frac{z_2}{k_2} = \frac{1}{k_1} + \frac{1}{k_2} - \frac{1}{|\mathbf{k}_1 - \mathbf{k}_2|}, \qquad (3)$$

and with

$$\Delta(\mathbf{k}_1, \mathbf{k}_2) = 2[(z_1/k_1) ln(k_1/\kappa) + (z_2/k_2) ln(k_2/\kappa)]. \qquad (4)$$

Both of these difficulties were ultimately overcome by the successful methods for treating the electron-impact ionization problem. The first of them, the asymptotic form in Eq.(1), was the central issue addressed by the Exterior Complex Scaling (ECS) method, which is the principal subject of this talk. The second of them, the Coulomb breakup amplitude formula in Eq.(2) and its attendant numerical pathologies, required a reformulation and the observation that numerical computations on a finite volume can be at most beset by a finite overall phase that leaves observables unaffected.

2 An Abridged History of Calculations of Differential Cross Sections for Electron-Impact Ionization

It is obviously not possible to do justice to a review of calculations, much less the theory, of electron-impact ionization that lead to today's state-of-the-art. A great deal is known about the fundamental theory, for example the $\sigma \sim E^{1.127}$ threshold law [6] and the asymptotics of the problem, but that rich literature is beyond the scope of this talk. Nonetheless, while a review of the theory of this subject is not possible here, it is useful in the context of the effort to reduce the (e,2e) problem to "practical computation" to note at least *some* of the general categories into which *computational* approaches to this problem have fallen.

Prior to 1987, all calculations of the most detailed cross sections for electron impact ionization, the triply differential cross section (TDCS) that is dependent on both the energy and the angles of the two outgoing electrons, were "perturbative" in nature. The breakup amplitude in these calculations takes the form $\langle \Psi^{(-)}_{approx} | V_a | \Phi_a \rangle$, where Φ_a is the initial state and $\Psi^{(-)}_{approx}$ is some approximation to the scattering wave function. For incident energies above several keV the Born approximation, most notably investigated by Inokuti

and coworkers [7], can describe the TDCS accurately. There is also an extensive literature on second [8] and higher order Born approximations [9].

A class of distorted wave Born approximations (DWBA) that describe the TDCS well at energies of 500 eV and upward, particularly for final states in which one electron emerges with most of the energy, replaces one of the plane waves of the Born approximation for the outgoing electrons with Coulomb distorted waves. Variants of the DWBA that involve distorted waves for both electrons and others that include initial state interactions have also been investigated. Madison and coworkers [10] performed early DWBA calculations, while other workers have investigated variants called Coulomb-projected Born [11,12] and the distorted-wave impulse approximation [13]. A notable success in the general class of perturbative approaches is an approach due to Pan and Starace [14,15], called the "distorted partial wave" method, which for the specific final state geometry that has the two electrons leaving with an angle relative to one another of $\theta_{12} = 180^0$, can give accurate results down to energies below 20eV.

In 1989 a more sophisticated class of "ansatz" wave functions was introduced into calculations of perturbative form by Brauner, Briggs and Klar [16] to explore the effect of the correct Coulomb asymptotic form for breakup. This was a landmark calculation that stimulated a range of other investigations focusing on the key question that still remained as a barrier to nonperturbative, *ab initio* calculations. That literature has been reviewed by Lucey *et al.* [17] and by Jones and Madison [18], with the general conclusion that while the asymptotics and final state interactions may be important, including them without a better treatment of the interaction region does not much improve matters.

An entirely different category of calculation seeks to apply close-coupling techniques which replace the correct Coulomb three-body boundary condition with "two-body boundary conditions" appropriate for ordinary inelastic scattering of electrons from atoms. A new wave of such *ab initio* calculations was initiated by Curran and Walters [19] in 1987 who pioneered the application of "coupled pseudostates" to the calculation of the TDCS for three-body breakup. The essential idea here is to discretize the continuum for one electron as a set of square-integrable "pseudostates", while applying ordinary, short range, scattering boundary conditions to the other. Total ionization cross sections had been computed in this way before [20,21] in early and therefore primitive calculations, but not the TDCS.

In this approach the channels in an ordinary close-coupling expansion consist of discrete states of the target (say, the hydrogen atom) plus a set of discrete approximations to continuum states of the target (square-integrable approximations to Coulomb functions). The other electron is treated as the scattered electron, whose channel eigenfunctions satisfy the usual boundary conditions appropriate to non-Coulomb scattering, $F_n(\mathbf{r}) \sim e^{i\mathbf{k}_0\cdot\mathbf{r}}\delta_{n0} + f_{n0}e^{ik_n r}/r$. The entire wave function, $\Psi^{(+)}$ is required to have the proper antisymmetry, although, for the necessarily finite number of pseudostates, the underlying dynamics in this approach treats the electrons nonequivalently. In the Curran and Walters calculations the breakup amplitude was then computed using an amplitude expression which ignores the formal considerations of Eq.(2), and is proportional to $\langle e^{i\mathbf{k}_f\cdot\mathbf{r}_1}\phi_{Coulomb}(\mathbf{r}_2)| - 1/r_1 + 1/r_{12}|\Psi^{(+)}\rangle$. These calculations gave encouraging preliminary results, but their primary impact was to be in the further refinement of these ideas.

The idea of coupled pseudostates was taken to its logical conclusion in the pioneering work of Bray, Stelbovics, Fursa and their collaborators in the form of the "convergent close-coupling" CCC method. By recasting the basic approach in terms of a coupled

Lippmann-Schwinger equation for the T-matrix in momentum space, and making use of large, orthogonal Laguerre basis sets, these workers pushed the notion of coupled pseudostates demonstrably to convergence – at least for the total ionization cross section. In this approach one must interpret the T-matrix elements for transitions to continuum pseudostates by providing a connection to the correct density of states in the continuum. Thus both the asymptotic boundary conditions of Eq.(1) and the amplitude expression of Eq.(2) are ignored in the CCC approach. Nonetheless, in a landmark paper that drove a wedge into the formal difficulties of the (e,2e) problem, Bray and Stelbovics [22] demonstrated that total ionization cross sections could be computed in an *ab initio* calculation to essentially arbitrary accuracy. The CCC method has been applied with success to the calculation of the TDCS for electron-impact ionization of hydrogen [23] as well as larger atoms in approximations with two active electrons. However the inequivalent treatment of the two electrons, and the fact that the formal boundary conditions for Coulomb breakup in Eq.(1) are ignored, lead to nonuniform convergence at the level of the single differential cross section [24,25].

In a related series of papers, Watanabe and coworkers [26,27] have applied R-matrix propagation to this problem, again applying two-body boundary conditions to the calculation of the breakup cross section. They have seen the same nonuniform convergence in the SDCS and have not treated TDCS to date. However, because of the efficient propagation scheme they employ, they have been able to do calculations covering a large region of space and therefore investigate the threshold region of the ionization process.

These questions have been the subject of a number of studies and call for approaches that either correctly incorporate the three-body Coulomb asymptotic boundary conditions or correctly avoid them within a rigorous formalism. One such approach is the "time-dependent close-coupling" approach of Pindzola, Schultz, Robicheaux and coworkers. In this approach a wave packet is fired at the target atom and the time-dependent Schrödinger equation describing its dynamics is solved in a close-coupling formulation [28,29]. There is thus no ambiguity about the boundary conditions, since the time-dependent Schrödinger equation is solved as an initial value problem. There is a price to be paid however, since the small mass of the electron causes the wave packet to spread radically during the time of the collision, making the analysis of the outgoing wave function the primary challenge. Nonetheless, this method has been shown to produce accurate total and single differential cross sections [30,31]. While TDCS calculations by this method are in progress and have not yet appeared, it is arguably the case that time-dependent methods scale better than most alternatives with increasing numbers of electrons, and that they may ultimately become the methods by which multiparticle breakup is first explored in accurate *ab initio* calculations.

The first method to successfully avoid the barrier of the three-body Coulomb boundary conditions in a formally correct manner and also address the issue of the extraction of the breakup amplitude from the wave function that solves the Schrödinger equation, was the method of Exterior Complex Scaling (ECS). In its first application [32] to the full problem of electron-impact ionization of hydrogen it was shown to provide quantitative agreement with experiment for the TDCS at low energies and to provide a complete framework by which the (e,2e) problem is reduced to "practical computation". It is to that method, the central subject of this talk, to which we now turn.

3 Exterior Complex Scaling and Reducing the e,2e Problem to Practical Computation

The method of exterior complex scaling and its applications have been developed in a series of papers [33,34,35,32,36,37,38,39,40], which from the outset divided the solution of the problem of electron-impact ionization into two discrete steps.

1. Compute the scattering wave function without recourse to the explicit three-body asymptotic form by applying exterior complex scaling to the solution of a discretized representation of the Schrödinger equation.

2. Extract differential and total ionization cross sections from the wave function by either "interrogating" it to compute the scattered flux, or using it in an integral expression for the breakup amplitudes.

3.1 Computing the Scattering Wave Function for Collisional Breakup

To begin the first step we must isolate the outgoing portion, Ψ_{sc}, of the full scattering wave function $\Psi^{(+)}$, because it is to that portion that Eq.(1) applies for breakup.

$$\Psi^{(+)} = \Psi_{sc} + \Phi_0. \tag{5}$$

where Φ_0 is the initial, unpreturbed state,

$$\Phi_0 = \frac{1}{\sqrt{k_0}}(e^{i\mathbf{k_0}\cdot\mathbf{r_1}}\varphi_0(\mathbf{r_2}) \pm \mathbf{e}^{\mathbf{ik_0}\cdot\mathbf{r_2}}\varphi_0(\mathbf{r_1})), \tag{6}$$

$\mathbf{k_0}$ is the incident electron momentum and φ_0 is the initial state of the atom. The scattered wave then satisfies the driven Schrödinger equation for a particular initial condition,

$$(E - H)\Psi_{sc} = (H - E)\Phi_0. \tag{7}$$

The ECS method owes its origins to the long history of complex scaling methods in atomic and molecular physics, which in turn is based on a very simple observation about the behavior of solutions of the Schrödinger equation when viewed as functions of complex variables. A purely outgoing wave, $\exp(ikr)$, with $k > 0$, becomes exponentially decaying when the coordinate, r, is scaled into the upper half complex plane, $\exp(ikre^{i\eta}) \to 0$ as $r \to \infty$. That observation applies to the asymptotic form in Eq.(1), in spite of its complicated logarithmic phases. Thus, complex scaling reduces the Coulomb boundary condition for breakup to the trivial condition that $\Psi_{sc}(\mathbf{r_1}, \mathbf{r_2})$ vanish at infinity.

The observation that complex scaling could simplify the boundary conditions for the case in which two electrons are in the continuum is due to Pont and Shakeshaft [41] in the context of double photoionization. Double photoionization is a "half collision" problem which admits to other simplifications as well, and there is at least one other method [42] that, making use of semiclassical outgoing wave boundary conditions, avoids the complications of the formal asymptotic boundary conditions in that context.

In the (e,2e) problem, however, we must extract the physics of breakup from Ψ_{sc} in a region in which the coordinates on which it depends are real, so we need to apply the complex scaling transformation only when either of the coordinates of the two electrons are greater than some radius, R_0. The ECS transformation that does this was invented and

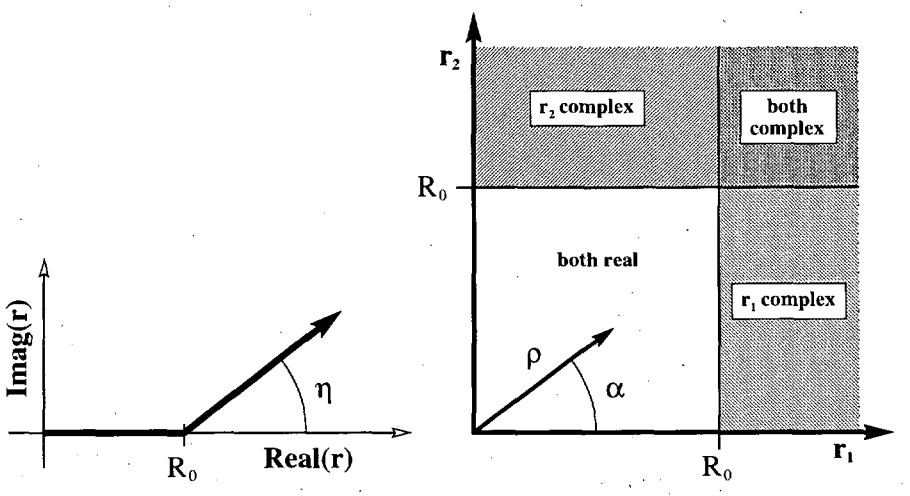

Figure 1. Left Panel: illustration of the ECS contour rotated into the upper-half of the complex r-plane beyond R_0. Right Panel: Depiction of exterior complex scaling for two radial coordinates.

investigated in the context of electron scattering resonances with only one electron in the continuum [43,44], and its adaptation to the (e,2e) problem shown in Fig.(1).

$$r \to \begin{cases} r & r < R_0, \\ R_0 + (r - R_0)e^{i\eta} & r \geq R_0. \end{cases} \tag{8}$$

The only part of the method for solving Eq.(7) remaining to be specified is the underlying representation. For all the ECS calculations to date, Ψ_{sc} is first expanded in coupled spherical harmonics of the angular coordinates of the two electrons. The resulting coupled radial wave functions, $\Psi^L_{l_1,l_2}(r_1, r_2)$, are computed on a finite difference or "discrete variable representation" grid. A typical calculation might have ~ 450 points in each radial dimension, and for a given total angular momentum, L, have of the order of 24 (l_1, l_2) angular momentum pairs. The time consuming step of the calculation, now a modest computation on a massively parallel supercomputer, is the solution of sparse linear equations of the order of five million.

One of the many radial functions contributing to Ψ_{sc} that result from such a calculation is shown in Fig.(2). In that figure one can see the outgoing flux in the discrete, inelastic channels going out near the axes, while the ionization flux goes out for large r_1 and r_2 in the structures resembling ripples from a pebble dropped in a pond.

3.2 Extracting the Cross Sections and Amplitudes from the Scattering Wave Function

We now come to the second major step of the ECS method. With the scattering wave function in hand we have the problem of extracting the information it contains about electron-impact ionization – with no hints from its numerical representation about how to distinguish the parts that correspond to Rydberg state excitation from those that correspond to ionization. It would appear that one would be forced after all to match to Eq.(1) at the edges of the "box" of hyperradius R_0 to extract the ionization amplitude.

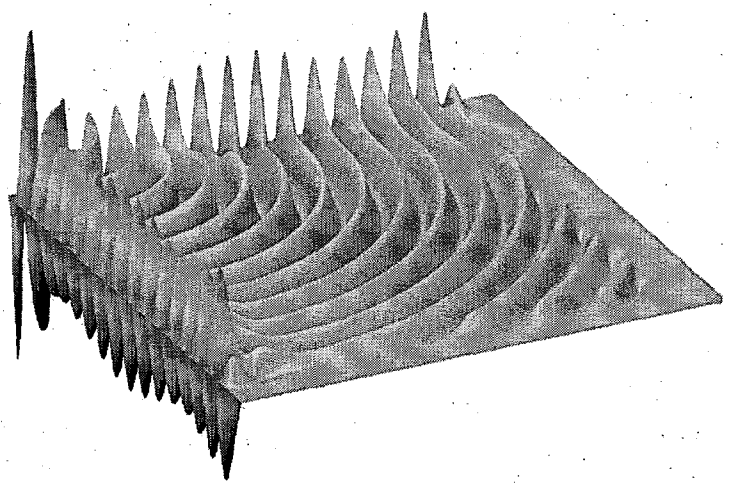

Figure 2. Real part of a representative radial function for electron-hydrogen scattering at 17.6 eV incident energy. Vertical axis is $Re(\Psi)$ and the two horizontal axes are r_1 and r_2 with origins at the rear left corner. $\Psi^L_{l_1,l_2}(r_1,r_2)$ is shown for singlet spin, $L = 2$ and $l_1 = l_2 = 1$

Initially, this problem was solved in the most primitive and straightforward way possible. One may simply compute the quantum mechanical flux,

$$\mathbf{F}(\rho,\Omega) = (1/2i)(\Psi^*_{sc}\nabla\Psi_{sc} - \Psi_{sc}\nabla\Psi^*_{sc}), \tag{9}$$

inside the region where the coordinates are complex, and by extrapolating to infinite grid sizes, separate the region where one electron's coordinate is large from the ionization region in which both coordinates are large. The first ECS calculations on electron-impact ionization of hydrogen were done in this fashion.

But ultimately it is advantageous, both computationally and theoretically, to confront the dilema posed by by the formal theory in Eqs.(2-4) . That was done in a study that first observed that the infinite phase that Eqs.(2-4) seek to avoid is simply an overall finite phase if the integration volume is finite. However, that is not the only pathology in those equations. The effective charges in the Coulomb functions that define the final state have the unfortunate property of destroying their orthogonality to the bound states of the hydrogen atom. In Eq.(2) there are terms that are proportional to momentum conserving delta functions if the integration volume is infinite, and which therefore contribute nothing to the ionization amplitude. The second observation, on which a practical and accurate formal expression for the breakup amplitude can be based, is that those terms also vanish on a finite integration region if the effective charges are set to unity.

The result is an integral expression that is appropriate to finite volumes,

$$f(\mathbf{k}_1,\mathbf{k}_2) = \langle\phi^{(-)}_{\mathbf{k}_1}\phi^{(-)}_{\mathbf{k}_2}|E - T + 1/r_1 + 1/r_2|\Psi_{sc}\rangle, \tag{10}$$

where $\phi^{(-)}_{\mathbf{k}_2}$ is a Coulomb scattering function. It is the application of this integral formula, together with the ECS method, to ionization of hydrogen that has given the most accurate description of the complete dynamics to date and which does in fact "reduce the problem to practical computation". We now turn to the results of these calculations.

4 Calculations on Electron-Impact Ionization of Hydrogen

The magnitudes and shapes of the singly differential cross sections at low energies give a particularly compelling demonstration of the ECS approach and make a satisfying connection with the semiclassical theories that have been applied to the threshold behavior of the ionization process. Fig.(3) compares the SDCS computed by the two methods described in Section 3.2 at incident energies from 15.4 eV (only 2 eV above the ionization threshold) to 54.4 eV. At lower energies the flux and integral formula methods for computing the SDCS disagree by as much as 10%, because the extrapolation of the flux becomes increasingly difficult as the energy is lowered. However no such difficulty affects the integral expression in Eq.(10). At very low energies the SDCS is almost flat and almost constant as a function of incident energy. If it were flat and constant it would correspond to a linear threshold law for the total cross section. In semiclassical calculations at the Wannier geometry with electrons exiting in opposite directions, Rost [45] predicted qualitatively the subtle departures from flatness as the SDCS turns from a "smile" at high energies to a nearly flat shape at energies near threshold.

The TDCS at 17.6eV incident energy is compared with the absolute experimental measurements of Röder *et al.* [15] in Fig.(4). The results are shown for the coplanar symmetric experimental geometry (which means that the incident electron and both exiting electrons lie in a plane, *and* the two exiting electrons have equal energy), with a fixed angle between the exiting electrons. There has been some question about about the internormalization of these measurements with others done holding the direction of one exiting electron fixed, but unpublished results in J. Röder's thesis have recently resolved that discrepancy [46]. From 19.6 eV to 30eV only relative measurements are available, but excellent agreement with them is attained in ECS calculations, as is demonstrated in Fig.(5). Complete presentations of ECS results on this system up to August of 2001 are given elsewhere [37,40].

4.1 Conclusion

The reduction of this problem to computation means that with accurate calculations we can now confidently explore the dynamics at geometries that have never been measured. A glimpse of what is now possible is given in Fig.(6), which shows only three frames of an animation of the TDCS for nonequal energy sharing that demonstrates in detail how the angular distributions change as the fraction of the ejection energy taken by one electron varies from zero to one. For this simple system it will shortly become a routine matter to computationally explore noncoplanar geometries together with with unequal energy sharing only a few volts above threshold. At this stage it would be useful to have a final, absolute, experimental benchmark for one or two such configurations.

While there are aspects of the problem of the collisional breakup of a system of three charged particles that are not yet solved, notably the problem of positron impact ionization where ionization competes with positronium formation, it can truly be said that, finally, the simplest e,2e problem has been "reduced to practical computation".

Acknowledgments

This work was performed under the auspices of the US DOE by the Lawrence Berkeley and Lawrence Livermore National Laboratories under contracts DE-AC03-76SF00098 and W-

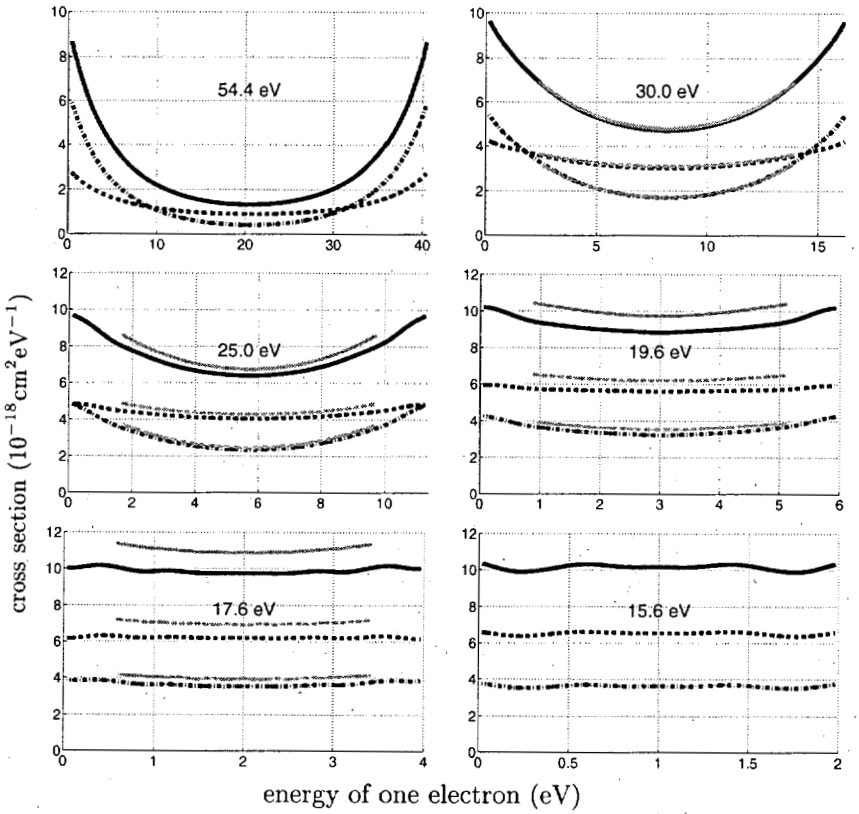

Figure 3. Singly differential cross sections for e-H scattering at the collision energies indicated. Individual components for singlet (dashed line) and triplet (dot-dash line) are shown. Where applicable, results based on flux-extrapolation are shown in light gray.

7405-Eng-48, respectively. The work was supported by the US DOE Office of Basic Energy Science, Division of Chemical Sciences, and computations were carried out at the National Energy Research Scientific Computing Center at Lawrence Berkeley National Laboratory.

References

1. E. A. Hylleraas, *Z. Phys.* **54**, 347 (1929).
2. C. L. Pekeris, *Phys. Rev.* **112**, 1649 (1958).
3. R. K. Peterkop, *Opt. Spectrosc.* **13**, 87 (1962).
4. M. R. H. Rudge and M. J. Seaton, *Proc. Roy. Phys. Soc. A* **283**, 262 (1965).
5. M. R. H. Rudge, *Rev. Mod. Phys.* **40**, 564 (1968).
6. G. Wannier, *Phys. Rev.* **90**, 817 (1953).
7. M. Inokuti, *Rev. Mod. Phys.* **43**, 297 (1971); M. Inokuti, Y. Itikawa and J. E. Turner, *Rev. Mod. Phys.* **50**, 23 (1978).
8. F. W. Byron Jr., C. J. Joachain and B. Piraux *J. Phys. B* **18**, 3203 (1985).

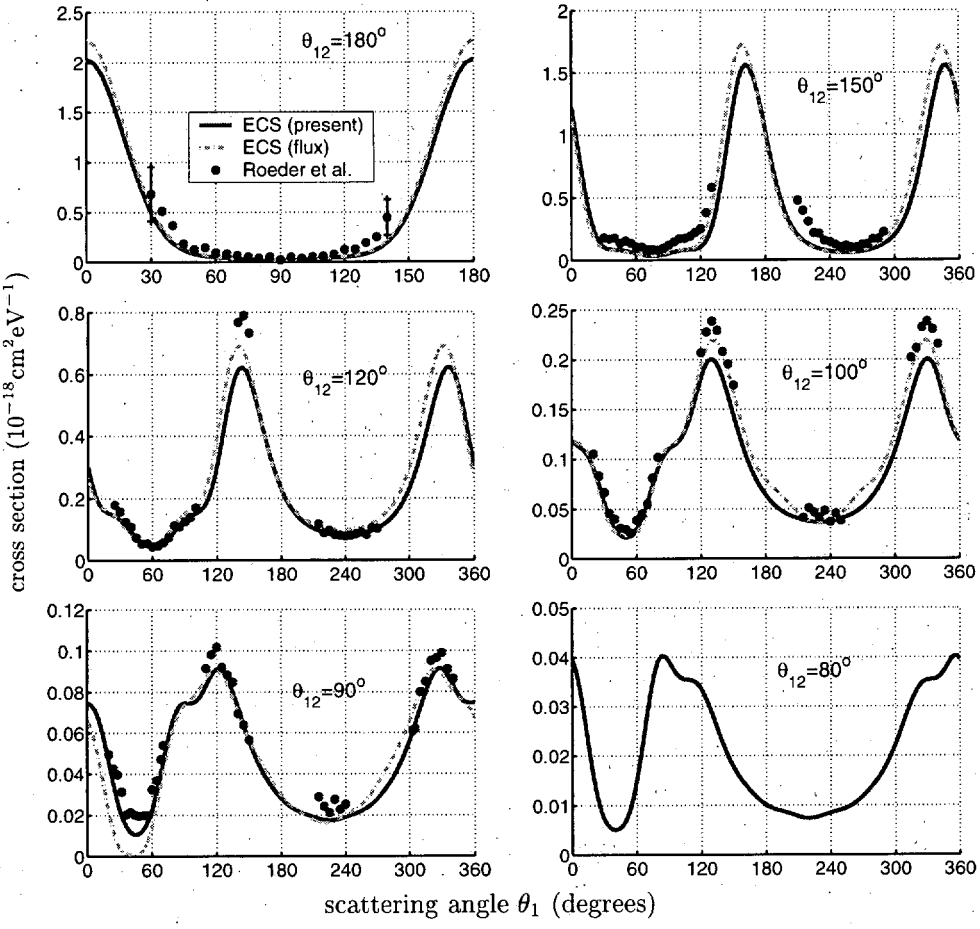

Figure 4. Equal-energy sharing, coplanar TDCS for 17.6 eV incident energy shown for geometries with θ_{12} fixed. Experimental data are absolute measurements of Röder *et al* with 40% error bars. Dark solid curves: integral expression for breakup amplitude, Lighter curves: flux extrapolation.

9. C. J. Joachain, B. Piraux, R. M. Potvliege, F. Furtado and F. W. Byron Jr. *Phys. Lett.* **112A**, 138 (1985).
10. D. H. Madison *Phys. Rev.* A **8**, 2449 (1973); D.H. Madison, R.V. Calhoun and W.N. Shelton, *Phys. Rev.* A **16**, 552 (1977).
11. S. Geltman and M. Hidalgo *J. Phys. B* **7**, 832 (1974).
12. C. T. Whelan, H. R. J. Walters, J. Hannsen and R. M. Dreizler, *Aust. J. Phys.* **44**, 39 (1991).
13. I. E. McCarthy and E. Weigold Phys. Rep. **27C**, 275 (1976).
14. C. Pan and A. F. Starace, *Phys. Rev. A.* **45**, 4588 (1992).
15. J. Röder, J. Rasch, K. Jung, C. T. Whelan, H. Ehrhardt, R. Allan, and H. Walters, *Phys. Rev. A* **53**, 225 (1996). J. Röder, H. Ehrhardt, C. Pan, A. F. Starace, I. Bray, and D. Fursa, *Phys. Rev. Lett.* **79**, 1666 (1997).

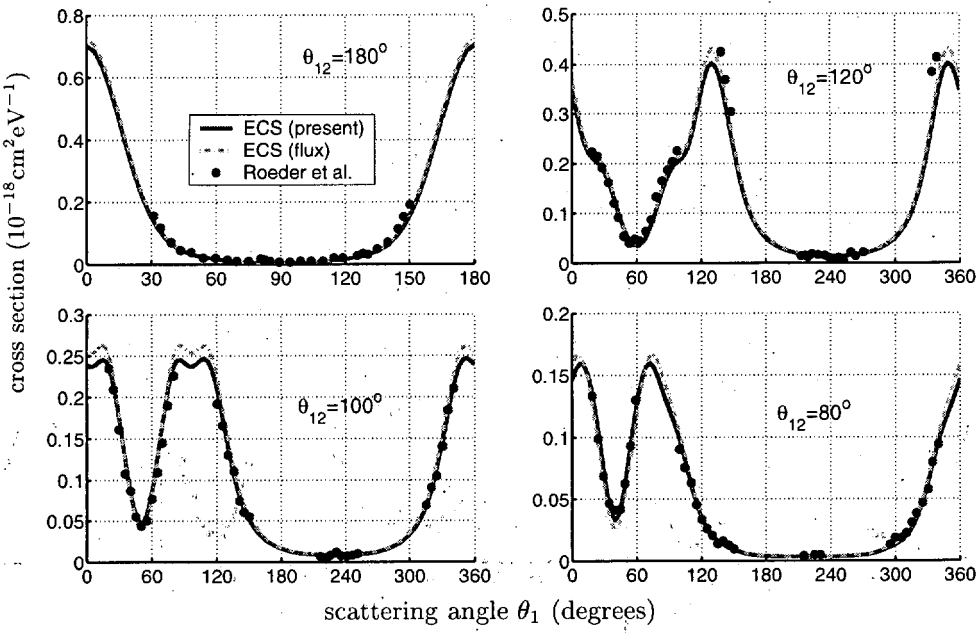

Figure 5. TDCS for 25 eV incident energy. Normalization factor to convert measured values of Röder *et al.* from arbitrary units is 0.16.

16. M. Brauner, J. S. Briggs and H. Klar *J. Phys. B* **22**, 2265 (1989).
17. S. P. Lucey, J. Rasch, and C. T. Whelan, *Proc. R. Soc. Lond. A* **455**, 349 (1999).
18. S. Jones and D.H. Madison, *Phys. Rev. A* **62**, 042701 (2000).
19. E. P. Curran and H. R. J. Walters *J. Phys.* B **20**, 337 (1987).
20. J. Callaway and D. H. Oza *Phys. Rev.* A **29**, 2416 (1984).
21. D. F. Gallaher *J. Phys.* B **7**, 362 (1974).
22. I. Bray and A. T. Stelbovics *Phys. Rev. Lett.* **69**, 53 (1992).
23. I. Bray, *J. Phys.* B **33** (2000).
24. I. Bray, *Phys. Rev. Lett.* **78**, 4721 (1997).
25. T. N. Rescigno, C. W. McCurdy, W. A. Isaacs and M. Baertschy *Phys. Rev.* A **60**, 3740 (1999).
26. D. Kato and S. Watanabe *Phys. Rev.* A **56**, 3687 (1997).
27. N. Miyashita, D. Kato and S. Watanabe *Phys. Rev.* A **59**, 4385 (1999).
28. M. S. Pindzola and D. R. Schultz *Phys.Rev.* A **53**, 1525 (1996).
29. M. S. Pindzola and F. Robicheaux *Phys.Rev.* A **54**, 2142 (1996).
30. M. S. Pindzola and F. Robicheaux *Phys.Rev.* A **55**, 4617 (1997).
31. M. S. Pindzola and F. Robicheaux *Phys.Rev.* A **61**, 052707 (2000).
32. T. N. Rescigno, M. Baertschy, W. A. Isaacs, and C. W. McCurdy, *Science* **286**, 2474 (1999).
33. C. W. McCurdy, T. N. Rescigno and D. Byrum *Phys. Rev.* A **56**, 1958 (1997).
34. C. W. McCurdy and T. N. Rescigno, *Phys. Rev. A* **56**, R4369 (1997).
35. M. Baertschy, T. N. Rescigno, W. A. Isaacs, and C. W. McCurdy, *Phys. Rev. A* **60**,

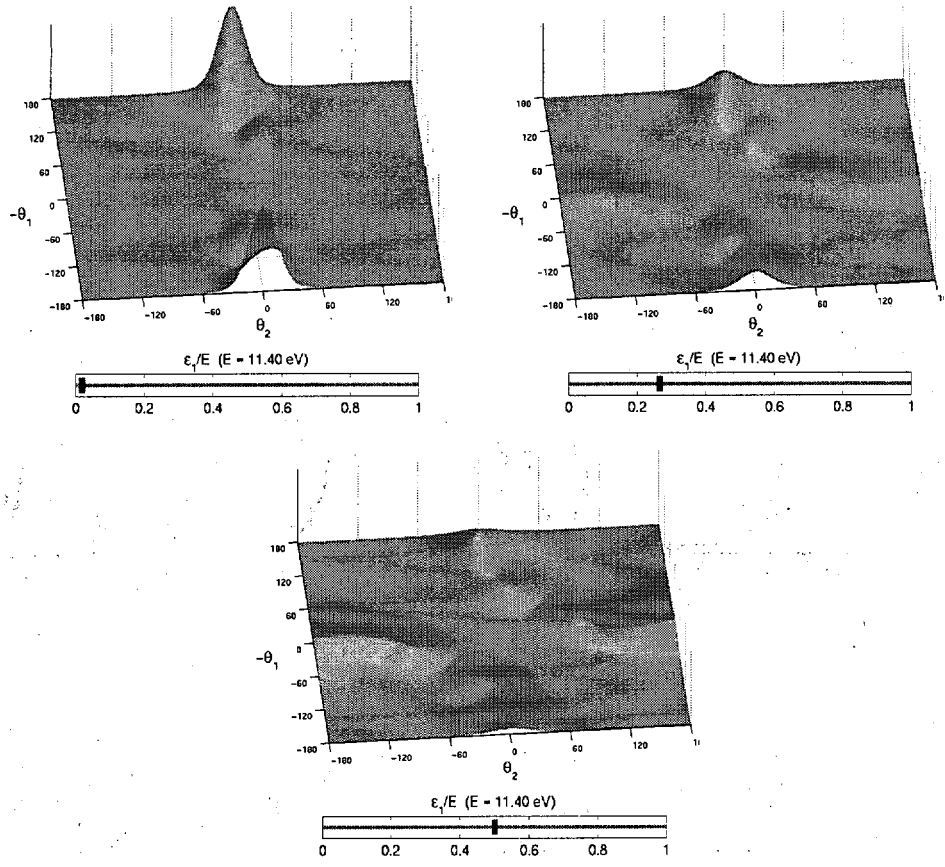

Figure 6. TDCS vs $-\theta_1$ and θ_2 for different energy sharings at 25 eV incident energy. Top left: $\epsilon_1 = 1/50E$. Top right: $\epsilon_1 = 1/4E$. Bottom: $\epsilon_1 = 1/2E$.

R13 (1999).
36. M. Baertschy, T. N. Rescigno, W. A. Isaacs, X. Li, and C. W. McCurdy, *Phys. Rev. A* **63**, 022712 (2001).
37. W. A. Isaacs, M. Baertschy, C. W. McCurdy, and T. N. Rescigno, *Phys. Rev. A* **63**, 030704R (2001).
38. C. W. McCurdy and T. N. Rescigno, *Phys. Rev. A* **62**, 032712 (2000).
39. C. W. McCurdy, D. A. Horner, and T. N. Rescigno, *Phys. Rev. A* **63**, 022711 (2001).
40. M. Baertschy, T. N. Rescigno, and C. W. McCurdy *Phys. Rev.* A **64**, 022792 (2001).
41. M. Pont and R. Shakeshaft *Phys. Rev.* A **51**, 494 (1995).
42. L. Malegat, P. Selles and A. K. Kazansky *Phys. Rev. Lett.* **85**, 4450 (2000).
43. B. Simon, *Phys. Letts.* A **71**, 211 (1979).
44. C. A. Nicolaides and D. R. Beck *Phys. Letts.* A **65**, 11 (1978).
45. J. M. Rost *Phys. Rev. Lett.* **72**, 1998 (1994).
46. J. Röder, *private communication.*